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Lead isotopes in soils near five historic American lead smelters and refineries

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Abstract

This survey of soil lead in the vicinity of old industrial sites examines how the stable isotope patterns vary among the sites according to the sources of the lead ore processed at each site. Lead smelters and refineries, which closed down decades ago, are the basis of this investigation. Samples were taken from near five old factory sites in Collinsville and Alton (Illinois), Ponderay (Idaho), East Chicago (Indiana) and Omaha (Nebraska). Historical records were searched for accounts of the sources of the lead. Lead concentrations were measured by atomic absorption flame spectrophotometry, and stable isotopic analysis was done by plasma ionization mass spectrometry. At every site visited, remnants of the old factories, in terms of soil lead pollution, could be found. In spite of potential complications of varying smelter feedstock sourced from mines of different geological age, it was possible to match the isotopic patterns in the soils with the documented sources of the ores. The Collinsville and Alton sites resembled Missouri lead. The Ponderay value was higher than major Bunker Hill, Idaho deposits, but closer to the minor, nearby Oreille County, Washington ores. Mostly Utah ore was used in East Chicago. The Omaha soil reflects lead from Mexico, Colorado and Montana.

Keywords: Soil lead; Stable isotopes; Smelters; History; Utah; Missouri; Nebraska; Illinois; Indiana; Idaho; Oklahoma; Washington; Refineries

Article Outline

1. Introduction
2. Area descriptions
 - 2.1. Collinsville, Illinois
 - 2.2. Alton, Illinois

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Reference #101

- 2.3. Ponderay, Idaho
- 2.4. East Chicago, Indiana
- 2.5. Omaha, Nebraska

3. Methods

4. Results

5. Discussion

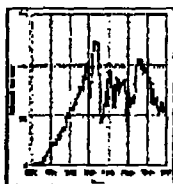
Acknowledgements

References

1. Introduction

Today, the American lead industry operates at only a fraction of its peak activity of many decades ago. Elevated soil lead levels are often to be found around former lead smelting and refining sites. This report examines how this excess soil lead isotopically resembles the historically documented sources of the lead ore processed at that particular factory site.

Current production of lead is approximately at levels last seen around 1910. Lead production peaked in the 1920s, with annual production of refined lead metal in the United States at nearly 750 kt, more than twice current levels (see Fig. 1). At that time, 21 primary smelters and refineries were in operation (Crane, 1927). Since then, the uses of lead in paint pigments and as a gasoline additive have been eliminated. Lead plumbing and cable covers have been largely replaced by other materials. Generally, there has been less demand for lead. The major use of lead today is in batteries, which use recycled scrap, rather than newly mined, lead. Production has continued only at larger and increasingly efficient and expensive factories. As a result, there are fewer operating smelters and refineries in the United States. There were 7 in 1973 and only 3 primary lead smelters and refineries operating in 1999 (Smith, 1999), producing about 350 kt/year. This tally is distinct from secondary smelters, which deal in scrap or recycled lead items. Those operations were much smaller and more numerous (Eckel et al., 2001).



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Fig. 1. Annual primary production of refined lead in the United States over the past 140 years. This lead is mostly from domestic mines, but imported ore and bullion refined in the United States are included. Scrap or recycled lead is not. Data are compiled from the US-Bureau of Mines Annual Minerals Yearbook.

The lead factories, which closed down decades ago, are the basis of this investigation. Historical records and field samples have been used to trace the lead from mines to smelter, where the ore is roasted driving off sulfur and converted to bullion, and onto a refinery for conversion of the bullion into lead metal (Hallows, 1943). This refined lead metal was used in making plumbing, electric cable covering, storage batteries and the manufacturing of white and red lead for paints and pigments and numerous other applications. Lead in its different grades of purity and forms was used in practically every community in the country (Crane, 1927).

Part of the rationale for this study is the wide variability of isotopic abundance ratios among the four stable isotopes of lead found in different ore bodies worldwide. Ore bodies which were formed at different geological times would have different amounts of primeval lead, present when the Earth formed, plus lead produced as an end-product of the radioactive decay of uranium and thorium over the history of the Earth up until the ore body

was formed. These isotopic patterns have been well studied and understood (Doe, 1970). These differences often allow lead to be traced and distinguished, which has proven useful in geochemistry, environmental health and archeology (Rabinowitz, 1995).

Table 1 shows the major lead producing states, their annual outputs during the peak years and the range of isotope ratios typically seen. The tonnage figures were compiled from the United States Bureau of Mines Annual Yearbooks. Although there were lead producing mines operating in more than 20 states, 3 states, Missouri, Idaho, and Utah, taken together, produced more than three-quarters of the lead ore.

Table 1.

Geographic sources of lead ores used by American smelters and their isotope ratios

| Mine location | Annual output kt/year 1925– 1935 | Range of $^{206}\text{Pb}/^{204}\text{Pb}$ ratios for most lead ores, excluding outliers |
|-----------------------------------|--|--|
| Missouri plus Kansas and Oklahoma | 298 | 20.0–22.7 |
| Idaho | 123 | 16.2–17.0 |
| Montana | 22 | 17.3–18.4 |
| Colorado | 22 | 17.9–18.2 |
| Utah | 167 | 17.9–18.6 |
| Mexico | 205 | 18.5–18.9 |

The tonnage figures were compiled from the United States Bureau of Mines Annual Yearbooks. The isotope data was compiled from Doe and Stacey (1974), Doe et al. (1979), Heyl et al. (1974), Leach et al. (1998), Stacey et al. (1968) and Zartman (1974).

The isotope data in Table 1 were compiled from the Lead Isotope Data Bank (Doe, 1992), Doe and Stacey (1974), Doe et al. (1979), Heyl et al. (1974), Leach et al. (1998), Stacey et al. (1968) and Zartman (1974). The full range of observed values for a state is not shown, but rather only the range of the major deposits is shown, excluding outlier values which may be of geological interest, but not representative of the bulk of the ore mined. Table 2 shows some illustrative examples of published values of lead isotope ratios for a variety of samples from these different states and Mexico. Within a single mining district and within a single mine, the isotope values do vary (Doe and Stacey, 1974), but general trends are apparent. Overall, larger deposits have less variable and lower $^{206}\text{Pb}/^{204}\text{Pb}$ ratios. Generally, the $^{206}\text{Pb}/^{204}\text{Pb}$ ratios of Missouri ores fall within a range of about 20–23, while for ores from Colorado, Utah and Montana the values fall between 17.5 and 18.5 (Cannon and Pierce, 1963, Doe et al., 1968, Graney et al., 1996, Sanford, 1992 and Unruh et al., 2000). Mexican ores have somewhat higher and Idaho ores lower $^{206}\text{Pb}/^{204}\text{Pb}$ ratios (James and Henry, 1993).

Table 2.

Some examples of published isotope ratios from major mining districts

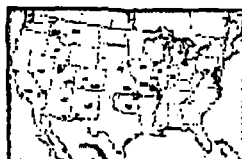
| Mining region | Sample and reference | Reported $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{206}\text{Pb}/^{207}\text{Pb}$ ratios |
|---------------|----------------------|---|
| Missouri | Herculeum MO Smelter | Rabinowitz 03 20.2 1.32 |
| | Doe Run Metal | Rabinowitz 03 20.8 1.34 |

| | | | | |
|----------|------------------------------------|---------------|-----------|-----------|
| | Galena KS slag | Rabinowitz 03 | 21.6 | 1.35 |
| | Picher OK ore | Cannon 63 | 22.0 | 1.37 |
| | Average SE Missouri ore | Heyl 74 | 20.8 | 1.30 |
| Idaho | Bunker Hill Mine, Coeur d'Alene | Leach 98 | 16.2 | 1.06 |
| | Star Mine, Coeur d'Alene | Leach 98 | 16.2 | 1.06 |
| | Average of 4 Bunker Hill Ingots | Rabinowitz 03 | 17.0 | 1.09 |
| | Average of 4 Pend Oreille WA mines | Small 73 | 17.4 | 1.11 |
| Montana | Emma Mine | Murthy 61 | 18.1 | 1.14 |
| | Lexington Mine | Murthy 61 | 18.0 | 1.14 |
| | Boulder Creek | Unruh 00 | 18.0 | 1.16 |
| | Anselmo Mine | Doe 68 | 17.9 | 1.15 |
| Colorado | Leadville/Gilman mine | Doe 74 | 17.8–18.2 | 1.14–1.17 |
| | Leadville mines | Graney 96 | – | 1.15–1.17 |
| | Lake City, Burrows Park | Sanford 92 | 18.2 | 1.17 |
| Utah | Tooele Smelter slag | Rabinowitz 03 | 17.9 | 1.15 |
| | Midvale Smelter soil | Rabinowitz 03 | 17.9 | 1.15 |
| | Bingham, Lark mine | Stacey 68 | 17.7 | 1.13 |
| | Park City, Apex mine | Stacey 68 | 17.9 | 1.14 |
| Mexico | Taxco ore | Cumming 79 | 18.7 | 1.20 |
| | Zacatecas ore | Cumming 79 | 18.8 | 1.20 |
| | Santa Eulalia ore | James 93 | 18.5 | 1.18 |
| | Tascatito ore | James 93 | 18.7 | 1.19 |

The other basis for this study is the long term persistence of lead pollution in soil (Chaney et al., 1989). Lead from factory operations, which falls on soil, is poorly soluble and will be retained for many centuries. Unless the site has been physically excavated or covered, lead added decades ago should still be present in the topsoil.

2. Area descriptions

Fig. 2 is an outline map of the United States showing the major lead mining districts and the location of the five smelters and refineries sampled. Major lead mining districts are shown in Southeast Missouri and the Missouri–Oklahoma–Kansas border, Colorado, Utah and in Northern Idaho. Smaller deposits were near the California–Nevada border, in Southern Arizona and New Mexico, Northeast Washington and along the Illinois–Wisconsin–Iowa border. Table 3 provides an overview of the five sites investigated, the corporate names and years of operation. Table 4 gives details of the soil sampling locations at each of these sites and the current land use.



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Fig. 2. Map of the United States showing the five industrial sites sampled (P for Ponderay ID, O for Omaha NE, A for Alton IL, C for Collinsville IL and E for East Chicago IN). Major lead mining districts are shown as shaded areas in Southeast Missouri and the Missouri–Oklahoma–Kansas border, two areas in Colorado centered in Durango and in Leadville, three close-by districts in Utah in Bingham, Park City and Tintic, and the Coeur d'Alene district in Northern Idaho. Smaller deposits were near the California–Nevada border, in Southern Arizona and New Mexico, Northeast Washington, and along the Illinois–Wisconsin border. The Tooele and Milford, UT smelter sites are shown with a T and M.

Table 3.

Five primary lead smelters and refineries sampled

| Location | Company name | Brand name | Operation | Years of operation | Years closed | Primary sources of ore |
|------------------|---------------|------------|------------------|--------------------|--------------|-------------------------------|
| Collinsville, IL | NL | St. Louis | smelter refinery | 1904–1938 | 66 | Missouri |
| Alton, IL | ASARCO | Federal | smelter refinery | 1902–1959 | 45 | Missouri |
| Ponderay, ID | Idaho S&R Co. | Idaho | smelter | 1907–1911 | 93 | Local mines including NE Wash |
| East Chicago, IN | ILR | Anaconda | refinery | 1912–1948 | 52 | Utah mostly, some Montana ar |
| | EP after 1948 | EP | refinery | 1948–1952 | | |
| Omaha, NE | ASARCO | O and G | refinery | 1899–1997 | 7 | Mix of Colorado, Utah, Montan |

ASARCO=American Smelting and Refining Co., NL=National Lead, ILR=International Lead Smelting and Refining Co., EP=Eagle Picher, O and G=Omaha and Grant.

Table 4.

Sample locations and current land use

| Location | ID # | Street address | Lat. °W | Long. °N | Date collected | Current land use |
|-------------------------|------|---------------------|---------|----------|----------------|-------------------|
| <i>Collinsville, IL</i> | | | | | | |
| | 386 | 1608 California | 38.6871 | 89.956 | June 03 | vacant lot |
| | 387 | Rotary Park | 38.6891 | 89.968 | | park |
| | 388 | 100 Pine Lake Rd. | 38.6904 | 89.961 | | old gate house |
| | 390 | 88 Pine Lake Dr. | 38.6917 | 89.969 | | home |
| <i>Alton, IL</i> | | | | | | |
| | 381 | by rail gate | 38.8751 | 90.140 | June 03 | industrial—closed |
| | 382 | west of Chesson Ln. | 38.8745 | 90.140 | | industrial—closed |
| | 383 | west of main gate | 38.8792 | 90.147 | | industrial—closed |
| | 384 | by main gate | 38.8786 | 90.146 | | industrial—closed |
| | 385 | end of Cut St. | 38.8780 | 90.145 | | industrial—closed |

Ponderay, ID

| | | | | | |
|-----|-------------------|---------|---------|---------|--------------|
| 396 | end of Cedar Ave. | 48.3013 | 116.531 | June 01 | recreational |
|-----|-------------------|---------|---------|---------|--------------|

East Chicago, IN

| | | | | | |
|-----|------------------|---------|--------|----------|-----------------|
| 498 | 4912 McCook Ave. | 41.6242 | 87.466 | March 00 | vacant lot |
| 499 | 427 E 149 St. | 41.6236 | 87.467 | | housing complex |
| 501 | Goodman Park | 41.6258 | 87.470 | | park |
| 502 | Goodman Park | 41.6261 | 87.469 | | park |

Omaha, NE

| | | | | | |
|-----|------------------|---------|--------|---------|--------------|
| 376 | 4050 Ave. F | 41.2674 | 95.914 | June 03 | home |
| 378 | Levee N of I 480 | 41.2621 | 95.919 | | conservation |
| 379 | Levee N of I 480 | 41.2617 | 95.919 | | conservation |
| 380 | Levee S of I 480 | 41.2590 | 95.919 | | conservation |

2.1. Collinsville, Illinois

The St. Louis Refining Company established its lead refinery in nearby Collinsville, Illinois in 1904. It soon became part of National Lead (NL), also known as the Lead Trust. The factory closed on November 21, 1933, following a labor strike. The equipment was dismantled and shipped to Argentina (Stehman, 1992).

The lead ore for this smelter came from Southeastern Missouri, across the Mississippi River. That area produced more than 40% of domestic lead ore during the twentieth century and remains productive today. Because most markets for lead metal at that time were to the east and because tolls across the river favored shipping ore east rather than shipping fuel for the smelters westward, this large operation was sited in Illinois, near coal mines, rather than near the lead mines. Zinc works, which were even more fuel dependent, had been at the site since 1873 (Norton, 1812).

These Missouri lead ores have low silver content, and so it is unnecessary to refine them extensively to extract the silver, in contrast to the complex ores more typically found in the Rocky Mountains where silver is a valuable by-product. For those complex ores, a multi-step metallurgical extraction process was required. However, the Collinsville factory, and the similar works in Alton, was combined smelter-refinery operations, since the refining part of the operation was relatively simple (Ellis, 1926).

Today, the Collinsville smelter site is in a residential area, a suburb of St. Louis, with well-kept, single family homes. Illinois and Federal officials have said topsoil may need to be removed to protect citizens. Slag piles were encountered as the site was developed for housing. The officials have focused on a 150 acre tract averaging 1300 µg/g, well above the 20–35 µg/g level taken to be natural and safe and above the current Federal Environmental Protection Agency guidance level for residential soils of 400 µg/g (Pierce, 2003).

2.2. Alton, Illinois

The Federal lead works in Alton further north along the Mississippi, and also in Madison County, was begun by American Smelting and Refining (ASARCO) in 1903. This site was chosen because it provided high ground to protect the smelter from river flooding and "plenty of swamp area adjoining to get rid of the slag quickly and cheaply" (Anon., 1936). Other industries were already well established in the town including glass and steel works. In 1913, a bath house and mandatory clothing changes were provided for worker health protection. After several expansions, by 1926 the Federal plant was the largest lead smelter in the world (Ellis, 1926). By 1936, 40 acres of swamp were 40 ft deep under slag, made at 200–500 t daily, amounting to 2.5 Mt since the plant began. For decades, the entire output of the plant was used for telephone cables. The smelter closed in 1959 when plastics replaced lead in the coating for wires and the plant switched to smelting aluminum before closing in 1981 (Klope, 1986). The location today is an unused, overgrown site within an area devoted to heavy industry, awaiting economic reactivation.

2.3. Ponderay, Idaho

By 1900 Idaho was a major lead, zinc and silver mining area, producing about 1/4 of all domestic lead. Mostly, it was along the Coeur d'Alene River. During the early history of this district, the ores were transported to ASARCO works in Leadville, Colorado (open since 1879), Selby, California (opened 1898) or Tacoma, Washington (opened 1896) for smelting and refining (Alken, 1993). Not until 1917 was the large Bunker Hill smelter and refinery built in Kellogg, Idaho. Several small smelters operated until then: Idaho Smelting and Refining Company in Ponderay from 1907 to 1911, the North Fork Smelting and Mining Company in Enaville from 1910 to 1911, the Red Bird Smelter Company in Clayton from 1912 to 1916, the Lemhi Smelter Company in Hahn from 1910 to 1911 and the Greyhound Company in Sea Foam in 1910. These small works all ceased as the smelting operations consolidated in Kellogg in order to handle the output from the productive Idaho mines. The Bunker Hill smelter operated until 1981. Of these several small operations, only Ponderay was visited.

Currently, the Ponderay smelter site is marked by a slag pile along the lake, which was the major means of area-wide transportation. Ponderay is about 80 miles from Kellogg. Old rails related to plant operations are still present, nearly a century later, on the top of the slag heap by the lake. During hot weather, this is a popular swimming and picnic spot in a residential area.

2.4. East Chicago, Indiana

This site is in an area of heavy industry. In 1911 International Smelting and Refining Company (ILR), a subsidiary of Anaconda Copper, constructed this refinery in East Chicago, Indiana, near Chicago, Illinois, to process lead bullion from its smelters in Tooele, Utah, and from the ASARCO smelter in East Helena, Montana. The Tooele bullion was cast in four-ton slabs and the East Helena bullion in 100-lb bars, both arriving by rail. The lead bullion was de-silvered and refined to make nearly pure lead anodes (Johnson, 1929). Some of these anodes were electrolytically converted to white lead (a hydrated lead carbonate) at the same East Chicago factory, for use as lead paint pigment, sold under the Anaconda label (Heckel, 1934). Zinc oxide, silver and antimonial lead were also produced, besides lead metal.

Large steel mills and chemical factories were in this area and the immediate neighborhood in East Chicago hosted other lead works. In 1910, US Smelting and Refining (USS) had built an electrolytic lead refining works, less than a mile away from the ILR factory. However, until it closed in 1972, during most of its life, it was a secondary smelter processing scrap lead metal, often from batteries. Only lead refined directly from lead ore, not secondary scrap lead, was used to make corroding grade lead, essential for the production of white lead paint. This high purity grade of lead metal was called corroding because most white lead was made by corroding the metal. For white paint to be satisfactory, it had to be free of impurities, which might affect the color, so this premium corroding grade of refined lead metal was manufactured. Within 10 miles of the ILR refinery in Calumet, Illinois, the large Sherwin-Williams Paint Company operated corroding sheds (1919–1953) where white lead was

made from lead metal. The Glidden Company operated a litharge and red lead pigment factory within 2 miles of the ILR refinery, in Hammond, which required lead. Many other customers for lead metal operated in this industrial area. Eagle Picher (EP) briefly owned the ILR refinery operation for 6 years before it closed in 1952 (Schulenberg, 1988).

Since 1968, when the ILR site was rehabilitated, including extensive soil excavation and replacement, it has been a Federal Housing and Urban Development housing complex. Today, the old ILR Anaconda factory site contains several apartment houses and a recreation building (the old zinc works). Additionally, a park with playgrounds was created from the Northern end of the factory site. The immediate surrounding neighborhood is economically depressed, with small multifamily housing.

The lead bullion for this refinery came largely from the ILR smelter in Tooele, Utah. Since the 1870s, lead ores from various mines in Utah were smelted in Salt Lake City, a valley. However, law suits by farmers forced smelters within Salt Lake City to close. So, in 1911, ILR began construction of the Tooele smelter in the next valley, to handle local ores from 15 mines in the nearby Bingham District, Park City and Tintic, and also some ore concentrates transported from Idaho and Nevada mines. The Tooele smelter closed in 1974, long after the East Chicago refinery closed (Dunlavy, 1986).

2.5. Omaha, Nebraska

The first lead refinery in Omaha, Nebraska was started in 1870, and by 1885 there was a white lead factory (Heckel, 1934). The large Omaha ASARCO plant was not built until 1899. It was named "Omaha and Grant" after its builder, James B. Grant, Later Governor of Colorado. It was a refinery, the smelting having been closer to the mine sites around the West. This placed the refinery on the rail lines, closer to the Eastern markets. It received lead bullion from widely spread ASARCO smelters: Leadville (1879–1961) and Durango, Colorado (1882–1938), Midvale, Utah (1905–1958), Chihuahua (1907–1938) and Coahuila (1913–1938), Mexico, El Paso, Texas (1886–1985), which also smelt Mexican ores along with some lead ore from Arizona and New Mexico, and East Helena, Montana (1889–April 2001). Of these, only the East Helena smelter was still operating when the Omaha refinery closed in December 1997. For those few years from 1997 to 2001, that bullion was sent to Mexico for refining. The Omaha refinery site itself was cleaned and decontaminated and, today, it serves as a park and restaurant. In November 1999, ASARCO was bought by Grupo Mexico S.A. de C.V. The plant emissions have impacted a large part of the city of Omaha, requiring remediation of 8840 acres with 66,515 residents and 20 public schools, because their soil lead levels exceeded 400 $\mu\text{g/g}$ (US EPA, 2002). The samples reported here were taken from just East across the river in Council Bluffs, Iowa, less than 1 km away.

3. Methods

Soil and slag samples were taken from the top 2 cm, typically five 20-g samples from over an area of 1 m^2 using a plastic scoop, choosing spots with minimal vegetation or influence by any nearby structure or road. These samples were dried, hand crushed, and coarsely sieved with a 1-mm mesh stainless steel screen. The lead was extracted using 10% nitric acid at room temperature for 2 days. Aliquots were diluted for atomic absorption flame spectrophotometry with background correction to determine the lead concentrations of the solutions. The results are expressed as $\mu\text{g Pb/g}$ dry soil material. When abnormally high values were found, for example, more than 1000 $\mu\text{g/g}$ at a site of an old lead factory compared to natural levels of less than 50 $\mu\text{g/g}$, and in the absence of other potential sources like highways, bridges, buildings and other industries, it is assumed that essentially all of the lead in the sample came from that factory.

These samples, thought to reflect the emissions from the factories, were then further diluted for isotopic analysis with a single collector, sector field mass spectrometer using plasma ionization (SFICPMS, Finnigan MAT model Element 1) by Lary Ball of the Woods Hole Oceanographic Institute. Quality assurance was addressed using

National Institute of Standards and Testing standard reference material #981 analyzed along with each batch of samples. Ten assays gave a mean value $^{206}\text{Pb}/^{204}\text{Pb}$ of 16.94 (S.D. 0.01), $^{206}\text{Pb}/^{207}\text{Pb}$ was 1.0933 (S.D. 0.0006) and $^{206}\text{Pb}/^{208}\text{Pb}$ was 0.4615 (S.D. 0.0002), which is indistinguishable from the certified values of 16.93, 1.0930 and 0.4613, respectively. The analytical uncertainty was much less than the range of values encountered.

4. Results

Table 5 contains the soil lead concentrations and isotopic abundance ratios and Fig. 3 provides a scatter graph of the isotope ratios. Only the samples with the highest lead concentrations at each site were submitted to isotopic analysis. At the Collinsville and Alton sites, similar patterns are apparent. High concentrations could be found, and the isotope ratios resemble Missouri lead, with $^{206}\text{Pb}/^{204}\text{Pb}$ ratios well above 20. These values are entirely consistent with the survey of Southeastern Missouri ores reported by Heyl et al. (1974). They reported an average of 131 ore samples of 20.8, ranging from 19.96 to 21.46 (Table 2).

Table 5.

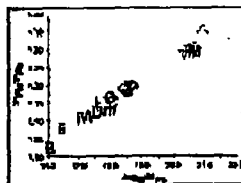
Soil sample lead data

| Location | ID # | Lead ($\mu\text{g/g}$) | Isotope | Abundance Ratio | |
|-------------------------|------|--------------------------|-----------------------------------|-----------------------------------|-----------------------------------|
| | | | $^{206}\text{Pb}/^{204}\text{Pb}$ | $^{206}\text{Pb}/^{207}\text{Pb}$ | $^{206}\text{Pb}/^{208}\text{Pb}$ |
| <i>Collinsville, IL</i> | | | | | |
| | 386 | 190 | 20.52 | 1.298 | 0.518 |
| | 387 | 117 | | | |
| | 388 | 12,740 | 20.92 | 1.352 | 0.560 |
| | 390 | 1,625 | 20.68 | 1.306 | 0.521 |
| <i>Alton, IL</i> | | | | | |
| | 381 | 17,202 | 20.52 | 1.297 | 0.519 |
| | 382 | 274 | | | |
| | 383 | 680 | | | |
| | 384 | 4,156 | 20.52 | 1.297 | 0.519 |
| | 385 | 2,454 | 20.34 | 1.287 | 0.516 |
| <i>Ponderay, ID</i> | | | | | |
| | 396 | >1,000 | 17.71 | 1.137 | 0.470 |
| <i>East Chicago, IN</i> | | | | | |
| | 498 | 1,360 | 18.59 | 1.191 | 0.484 |
| | 499 | 545 | 18.66 | 1.192 | 0.485 |
| | 501 | 430 | 18.49 | 1.188 | 0.485 |

| | | | | |
|-----|-----|-------|-------|-------|
| 502 | 475 | 18.50 | 1.190 | 0.485 |
|-----|-----|-------|-------|-------|

Omaha, NE

| | | | | |
|-----|-----|-------|-------|-------|
| 376 | 80 | | | |
| 378 | 441 | 18.51 | 1.187 | 0.485 |
| 379 | 164 | 18.69 | 1.197 | 0.485 |
| 380 | 112 | | | |



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Fig. 3. Scatter plot of lead isotope ratios. The soil samples from the five sites are triangles pointed up and down for Collinsville and Alton, the circles for Omaha, the diamonds for East Chicago and the square for Ponderay. Major mining districts are shown as letters, B for Bunker Hill, U for Utah, m for Montana, C for Colorado, X for Mexico and M for Missouri. The small l's represent four Idaho ingot samples from the Bunker Hill refinery, and the small o is for an Omaha and Grant ingot.

The Ponderay value of 17.7 was nearly exactly the same as what we found for one of the Bunker Hill ingots, cast in 1950 (Rabinowitz, 2002), although higher than the values seen in the large Coeur d'Alene lead ore deposit, which are closer to 16.2 (Leach et al., 1998). This indicates that most of the lead ore came from outside the extensive Coeur d'Alene deposits, which are about 80 miles away. More likely, the ore came from minor deposits from nearby Pend Oreille County, Washington, only 30 miles away by water, which are reported to average 17.4, ranging from 16.5 at the Newport mine to 18.8 at the Halfmoon Lake mine (Small, 1973).

The soil isotope values from East Chicago, with a $^{206}\text{Pb}/^{204}\text{Pb}$ near 18.5, are not consistent with only Utah and Montana ores as sources, but rather could be accounted for by a mixture of about four parts lead from Utah with a value less than 18, or Montana with $^{206}\text{Pb}/^{204}\text{Pb}$ values near 18, with about one part lead from Missouri, which has a ratio above 20 (Table 2). In that, Missouri was the largest lead producer and was much closer than Utah to East Chicago, and Eagle Picher (EP) had large lead interests in Southwest Missouri Tri-State area, this addition of Missouri lead would not be surprising.

The Omaha soil $^{206}\text{Pb}/^{204}\text{Pb}$ values near 18.5 or 18.6 are not far from the one Omaha and Grant ingot sample reported as 18.1 (Rabinowitz, 2003). Over its decades of operation, that refinery received varying amount of lead from smelters in Mexico, Colorado, Montana, Texas and Utah depending on tariffs and mine outputs, so its isotope value would not stay constant but would vary over a range. A similar pattern was noted at the Selby, California refinery where sources of lead ores varied over the many decades of operation and a particular batch of metal was isotopically different from accumulated factory effluent seen in nearby soils (Rabinowitz and Wetherill, 1972).

5. Discussion

In a previous report (Rabinowitz, 2002) on variations of the isotopic composition of different American brands of

corroding grade lead, the purity grade used to make lead paint pigments, data from lead metal products and also some soil were presented. Those represented metal samples from five brands (Bunker Hill, Doe Run, Eagle Picher, Omaha and Grant, and Selby) and soil values from four brands (Doe Run, Eagle Picher, International Lead Refining, Perth Amboy and USS Electro). In this survey, we provide soil data for five additional lead smelters-refineries, one of which, Omaha and Grant, we previously had reported a metal value. Taken together, all of the major primary smelters and refineries have been examined.

In reviewing the isotope ratio results from these five sites, considerable variation can be seen (Table 5). The two lead refineries in Illinois both had contaminated soils, which resemble nearby major Missouri lead ores (Table 2 and Fig. 2 and Fig. 3). The Ponderay, Idaho site did not resemble lead from the major Idaho lead mining district near Coeur d'Alene, but likely came from closer minor deposits in Pend Oreille County, Washington. The East Chicago ILR Anaconda lead refinery soil resembled a mixture predominantly of Utah and Montana lead ores, but it also had some lead from Missouri. The soils from near the Omaha refinery reflect the different mixtures of lead bullion, from Utah, Mexico, Colorado and Montana ores, which were processed there.

The different factories are not each isotopically unique (Table 5). That is, the ranges of values of some of the smelters do overlap. The systematic isotopic variation among the refineries is the result of lead ores of different geological ages being used. Assigning potential sources of lead ores to a smelter soil is necessarily inexact because of the variation of isotope ratios within a single mining district. No single number can be used to describe fully the ore from an area; rather a range of possible values is appropriate. So, even if isotope ratios in all the soils around a factory site were exactly the same, exact apportionment among potential sources is not possible.

This is a small study in terms of the number of soil samples examined at each site. For example, only one sample was analyzed from Ponderay. Therefore, generalizing about the whole history of a factory based on a single, perhaps unrepresentative, slag sample would be inappropriate.

Because the economic activity of bringing lead from afar to a factory can distort the natural isotopic composition in its vicinity, the opportunity arises to distinguish this anthropogenic lead from surrounding ambient lead. This ability to use lead isotopes as an inherent tracer to follow the dispersion and uptake of lead has proven useful. As one example, Gulson et al. (2004) examined teeth and environmental samples from an Australian community with a primary zinc-lead smelter. From these, they could estimate the impact of the smelter. That research team examined blood and milk from women who had moved before pregnancy from an area of different prevailing lead isotopic abundances, allowing them to observe the movement of skeletal lead during pregnancy and lactation (Gulson et al., 1999). Similar technology was used by Marskill et al. (1996) to measure the very slow vertical movement of lead in different types of soils at Roman and Mediaeval smelters in Britain. Hansmann and Köppel (2000) could distinguish between lead in Swiss soils that came from coal fly ash and lead from gasoline additive using this stable isotope method. Under the right conditions, stable isotopes provide a useful tracer or source identification technique (Rabinowitz, 1995). In this study, no sampling of the unpolluted or background ambient lead isotope ratios around these factory sites was performed. So, it is not possible to say if any of these sites would provide isotopically anomalous lead, compared to each one's own surroundings, and hence be potentially useful as tracers.

It is noteworthy that, although these industrial sites have been dormant for many decades, soil samples with elevated lead levels could easily be obtained. This reflects the immobility and persistence of lead in soil (Chaney et al., 1989). The isotopic patterns of soil lead are consistent with the documented sources of the ores used by each refinery.

Acknowledgments

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